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Serial No. 09/700,367

KARER et al.

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with a view to the teaching of Jorgensen et al. the Examiner acknowledged that the reference failed to specifically disclose applicants' requirements pertaining to the optional gas distributor plate which is situated in the region of transition of the reactor defined in Claim 1 and further specified in the dependent claims. The Examiner asserted, however, that a person of ordinary skill in the art would have been motivated to modify the percentage of the gas distributor plate surface which is occupied by orifices in the manner necessary to arrive at applicants' requirements because Jorgensen et al. state that a variation of the openings of the primary grid results in an adjustment of the pressure drop caused by the grid. The Examiner, therefore, concluded that the modifications which distinguish applicants' reactor arrangement from the reactor design taught by Jorgensen et al. were merely optimum workable conditions determined by a routine optimization of a result effective variable.

It is respectfully urged that the Examiner's conclusion is in error for failing to duly consider further pertinent information which is provided by the teaching of Jorgensen et al. and/or part of the background knowledge available to a person having ordinary skill in the art pertaining to gas-phase fluidized bed reactors. With regard to the latter, applicants herewith enclose copies of pages from a textbook on "Fluidization Engineering."1)

It is well known in the pertinent art that a pressure drop of at least 15%, preferably at least 30%, of the total pressure drop over the fluidized bed is necessary to achieve equal flows over the whole fluidization grid. This fact is, for example, corroborated by Jorgensen et al.'s statements in col. 5, indicated lines 46 to 54, of US 6,113,862, and by the explanations on page 102 of Kunii et al. It is also well known in the pertinent technology that the prerequisite pressure drop for fluidization is achieved by a fluidization grid having less than 10% of its surface taken up by openings, 2) corresponding to the statements of Jorgensen et al. that the primary fluidization grid has to obstruct at least 75% and preferably at least 90% of the area which is available for flow. 3) Essentially, a higher the obstruction which is provided by the fluidization grid provides for a higher pressure drop and vice versa, and a certain pressure

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<sup>1)</sup> Kumii et al., Butterworth-Heinemann, Newton, MA, 2<sup>nd</sup> Ed, 1991, pages 95 to 97 and 102 to 106; copy enclosed.

<sup>2)</sup> Cf., e.g., Runii et al., page 105.

<sup>3)</sup> Cf. col. 4, indicated lines 20 to 23, of US 6,113,862.

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drop is necessary to achieve the equal flows over the whole fluidization grid which are required for proper fluidization.

In light of the teaching of Jorgensen et al. and the technical background knowledge, any optimization of the percentage of openings in the surface of the fluidization grid would therefore be sought among grids which provide an obstruction of more that 75% or even more than 90% of the area in order to ensure the pressure drop which is necessary for proper fluidization and to achieve equal flows over the whole fluidization grid. Turning away from these principles and reducing the obstruction, ie. choosing to employ no gas distributor plate or a gas distributor plate which obstructs less than 50% of the area according to applicants' invention can, therefore, not reasonably be considered as a mere routine optimization of a result effective variable.

For completeness sake it is acknowledged that the secondary grid of the reactor addressed in the teaching of Jorgensen et al. may have an impact on the overall pressure drop in the reactor. However, bearing in mind that the secondary grid which is employed by Jorgensen et al. is preferably mesh like as depicted in Figure 2, the pressure drop contributed by the secondary grid is significantly lower than the pressure drop provided by the primary grid. The pressure drop over two grids can be calculated by simply adding the pressure drops over the grids, and any contribution to the overall pressure drop which is provided by the secondary grid is, therefore, negligible.

The foregoing shows that the Examiner's position that applicants' requirements are the result of a routine optimization of a result effective variable is not well taken. As pointed out in applicants' previous papers: obviousness under Section 103(a) requires that the motivation to make the changes which are necessary to arrive at the elements of the claimed invention and the reasonable expectation of success be found in the prior art, and neither the motivation nor the expectation of success can be based on the applicant's disclosure. Accordingly, the teaching of Jorgensen et al. cannot reasonably be considered to render applicants' invention prima facie obvious.

Favorable reconsideration of the Examiner's position and withdrawal of the rejection of Claims 1 to 4, 6 and 10 based on the teaching of Jorgensen et al. is therefore respectfully solicited.

<sup>4)</sup> Cf. col. 6, indicated lines 3 to 5, of US 6,113,862.

<sup>5)</sup> In re Vaeck, 947 F.2d 488, 20 USPQ2d 1438, 1442 (Fed. Cir. 1991).

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KARER et al.

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The foregoing explanations and arguments are also fully applicable where the Examiner's rejection of Claims 7 and 8 is concerned. The respective claims address an embodiment of the reactor defined in Claim 1 which further comprises a closable flap and the Examiner applied the disclosure of Lubbock for showing a slide valve, considered to be an equivalent to a flap. The disclosure of the secondary reference does not add anything to the teaching of Jorgensen et al. which would close or even narrow to gap to applicants' reactor as defined in Claim 1, and the respective requirements are incorporated into applicants' Claims 7 and 8 by reference. Obviousness under Section 103(a) relates to "the invention as a whole", and the question whether the disclosure of Lubbock can be taken to suggest the use of a closable flap is deemed to be moot in light of the differences between the elements of applicants' reactor which are specified in Claim 1 and the reactor which is addressed in the teaching of Jorgensen et al. It is therefore also respectfully requested that the rejection of Claims 7 and 8 based on the teaching of Jorgensen et al. and the disclosure of Lubbock be withdrawn. Favorable action is solicited.

Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees, to Deposit Account No. 14.1437. Please credit any excess fees to such deposit account.

Respectfully submitted,

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Encl.: Kunii et al., "Fluidization Engineering" Butterworth-Heinemann, Newton, MA, 2nd Ed, 1991, pages 95 to 97 and 102 to 106

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# Fluidization Engineering

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PREFACE PREFACE

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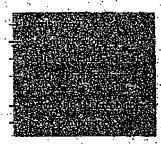
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The Dense Bed: Distributors, Gas Jets, and Pumping Power.

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This chapter focuses on what happens at the bottom of a dense fluidized bed and on the proper introduction of gas feed. We consider various distributor designs, their accompanying gas jetting problems, and the role of nozzles and their large gas jets as a means of promoting the circulation of bed solids. The chapter ends with a discussion of design procedures for distributors and of the pumping power requirement to keep the bed fluidized.

# istributor

### Ideal Distributors

Most small-scale studies in fluidization use ceramic or sintered metal porous plate distributors, because they have a sufficiently high flow resistance to give a uniform distribution of gas across the bed. This situation is ideal. Many other materials can do this—for instance, filter cloth, compressed fibers, compacted wire plate, or even a thin bed of small particles. Of course, some of these materials should be reinforced by sandwiching between metal or wire plates with large openings.

Although gas-solid contacting is superior with such distributors, for industrial operations they have several drawbacks:

- High-pressure drop leads to increased pumping power requirements; often a major operating cost factor:
- Low construction strength, hence impractical for large-scale use.
- High cost for some materials.
- Low resistivity against thermal stresses.
- Possible gradual clogging by fine particles or by products of corrosion.

Despite these disadvantages, compacted wire plates or sandwiched beds of small particles are sometimes used.

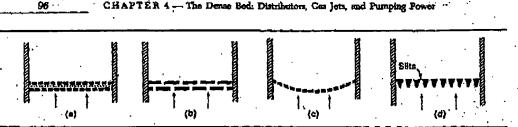


FIGURE 1
Plate and grate distributors are cheep and easy to construct: (a) sandwiching perforated plates;
(b) staggered perforated plates; (c) dished perforated plate; (d) grate bars.

### Perforated or Multiorifice Plates

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Perforated plate distributors are widely used in industry because they are cheep and easy to fabricate. Figure 1 illustrates several variations of a simple perforated plate distributor. Type (a) consists of two perforated plates sandwiching a metal screen that prevents solids from raining through the orifices when the gas flow is stopped. A variation of this, type (b), uses two staggered perforated plates and no screen.

One problem with this design is lack of rigidity. Large perforated plates deflect unpredictably under heavy load; hence, they need reinforcing for support. In addition, during thermal expansion gas leakage at the bed perimeter is possible.

When it is impractical to have a reinforcing structure to support a flat perforated plate against heavy loads, curved plates, such as type (c), are sometimes used. Curved plates will withstand heavy loads and thermal stresses. Because bubbling and channeling tend to occur preferentially near the center of a finidized bed, design (c) helps to counter this tendency. Distributor plates curved upward achieve good contacting only with more orifices near the perimeter and fewer near the center, a disadvantage for fabrication. Alternatively, parallel grate bars, type (d), may be used. These bars may be considered two-dimensional versions of perforated plates, and they have only seen limited use; see Fig. 2.23(b).

In some operations, large amounts of solids enter the bed with the tologous, for example in Exam's model IV FCC reactor, or in the multistage fluidized limestone calciner. In these situations perforated plates without screen are recommended.

The diameter of orifices in perforated plate distributors may range from to 2 mm in small experimental beds to as much as 50 mm in large FCC with their solid-entrained gases.

### Tuyeres and Caps

Perforated plate distributors cannot be used under severe operating conditions such as high temperature or a highly reactive environment. Tuyers designs (§ 2) are used in these situations. The multiple porous plate, type (a), gives (§ 2) gas distribution above each filter, but particles will settle between adjustments. Also, special precautions must be taken to ensure that the incoming is free of filter-clogging material. Types (b), (c), and (d) are frequently used prevent solids from falling through the distributor. However, with all [1]

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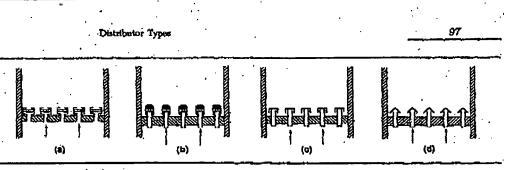
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FIGURE 2
Tuyere distributors: (a) porous plate type; (b) nozzie type; (c) bubble cap type; (d) sit nozzie type.

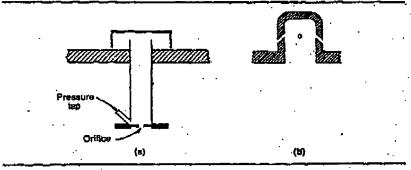


FIGURE 3 Details of tuyeres: (a) tuyers with inflow critics to control the gas velocity of gas extiling into the bed; (b) cap type.

designs, particles are apt to settle, sinter, and stick on the distributor plate itself. A variety of designs have been proposed and used to minimize this effect.

To ensure equal gas flow through the tuyeres of type (b), (c), or (d), each tuyere may be fitted with a high-resistance orifice at its gas inlet, as shown in Fig. 3(a). The cap-type tuyere of Fig. 3(b) has no orifice at its gas inlet. Instead, the orifices around the cap are designed to create a sufficient pressure drop for uniform fluidization. A disadvantage of this design involves the jetting effect of the high-velocity gas issuing from the orifices. This can cause considerable particle attrition. Conversely, the velocity of the gas issuing from the tuyeres of Fig. 3(a) can be chosen as desired because the rate of gas flow is fixed by the high-resistance inlet orifice. Because of their complicated construction, tuyere-type distributors are much more expensive than perferated plate distributors.

# Pipe Grids and Spargers

Experience shows that internals, such as properly placed heat exchanger tubes, substantially improve gas-solid contacting by breaking up growing bubbles and by preventing gulf streaming, or gross circulation of solids. In fact, proper design of internals can improve the quality of fluidization so much that refined high-resistance distributors are not needed. In such cases, a pipe grid or sparger, such as shown in Fig. 4(a), may be all that is needed to introduce reactant gas into beds fluidized by a second carrier gas coming from below. This is the

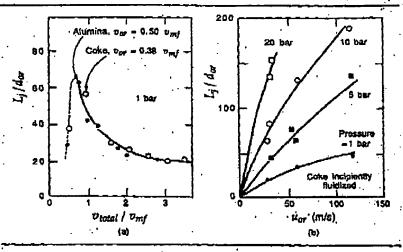
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CHAPTER 4 - The Deuse Bed: Distributors, Gas Jets, and Pumping Power



Effect of background flow and pressure upon the penetration depth of a vertical 1.55 mm; adapted from Yates et al. [5].

They correlate their findings for jet penetration length as follows:

$$\frac{L_{1}}{d_{\rm or}} = 21.2 \left(\frac{u_{\rm ox}^{2}}{g d_{\rm p}}\right)^{0.37} \left(\frac{d_{\rm or} z_{\rm ox} \rho_{\rm g}}{\mu}\right)^{0.05} \left(\frac{p_{\rm g}}{\rho_{\rm s}}\right)^{0.68} \left(\frac{d_{\rm p}}{d_{\rm or}}\right)^{0.24}$$

Equation (2) is just one of the many correlations proposed for jet length As with this equation, all other investigations have presented their findings in terms of  $L_i/d_{ox}$ . Massimilia [4] tabulates and then compares these findings in two diagrams,  $L_l/d_{or}$  versus  $u_o$  and  $L_l/d_{or}$  versus operating pressure, and find disagreement up to a factor of 100 or more. Since many of the pertinen variables, such as solid properties, size, and orifice diameter, differ from study t study, this is not surprising.

In conclusion, to predict the jet penetration length for a perticula application, choose the correlation for conditions that most closely match the system at hand and apply that correlation with caution.

Pressure Drop Requirements across Distributors

Experience shows that distributors should have a sufficient pressure drop  $\Delta p_z$ achieve equal flows over the entire cross section of the bed. According Zuiderweg [14], in the early years of fluidization engineering rules of the were followed, such as

$$\Delta p_d = (0.2 - 0.4) \Delta p_b$$

where  $\Delta p_{\rm b}$  is the pressure drop across the bed, given by Eq. (3.17). It is clear that increased Ap, will ensure a more even distribution of entering. However, an excessive  $\Delta p_d$  has its drawbacks.

 Power consumption and construction cost for the blower or comp increases with the total pressure drop, or  $\Delta p_t = \Delta p_b + \Delta p_d$ , and  $\Delta$ represent a significant portion of the total pressure drop.

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Pressure Drop Requirements across Distributors

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• For tayere distributors without inlet orifices (see Fig. 3(b)), the required distributor pressure drop to satisfy Eq. (3) may require the use of excessively high gas velocities at the nozzles. This may result in erosion and breakage of particles and an undesirable shift in size distribution of bed solids.

It is important, therefore, to know the minimum  $\Delta p_d$  that would ensure uniform fluidization in the required range of operations. From orifice theory and fixed bed equations, we can show that

$$\Delta p_d \propto u_0$$
 for porous plates (4a)

Several papers have published recommendations for relating  $\Delta p_d$  with  $\Delta p_b$  for satisfactory operations [15–21]. Hiby [15] lowered the gas flow to a portion of a bed and considered the bed stable if this caused  $\Delta p_b + \Delta p_d$  to decrease in this zone. From these experiments Hiby recommends that for stable operations, one should have

$$\frac{\Delta p_d}{\Delta p_b} = 0.15 \qquad \text{for } \frac{u_o}{u_{cof}} = 1-2 \tag{5a}$$

$$\frac{\Delta p_{cl}}{\Delta p_{b}} = 0.015 \qquad \text{for } \frac{u_{c}}{u_{mf}} > 2 \tag{5b}$$

Considering the channeling that may result from perturbations in a bed at close to  $u_{\rm mf}$ . Siegel [16] came up with the following criterion for stable operations:

$$\frac{\Delta p_d}{\Delta p_b} \ge 0.14 \tag{6}$$

Extending this channeling model, Shi and Fan [21] conclude that one can guarantee full fluidization if

. 
$$(\Delta p_d + \Delta p_b)_{\text{at eny } u_o} \cong (\Delta p_d + \Delta p_b)_{\text{at } u_{\text{ref}}}$$
 (7)

where, at umf.

$$\frac{\Delta p_d}{\Delta p_b} > \begin{cases} 0.14 & \text{for porpus plates} \\ 0.07 & \text{for perforated plates} \end{cases}$$
 (8a)

Finally, for hubbling beds of fine particles, meaning  $u_0 > u_{\rm mb}$  for Geldari A particles, supported on porous or perforated plate distributors. Mori and Moriyama [17] considered the consequences of shutting off the flow to part of the bed. With resumption of flow would the slumped solids refluidize? The result of their analysis suggests that for full fluidization one should have

$$\frac{\Delta p_d}{\Delta p_b} = \left(\frac{L_f}{L_{mf}} - 1\right) \frac{1}{1 - (u_{mf}/u_o)^n} \tag{9}$$

where n=1 for porous plates, n=2 for perforated plates. This expression shows that  $\Delta p_d$  must be large when operating close to  $u_{mf}$ , but can be lower when the bed operates at high  $u_0$ .

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CHAPTER 4 - The Dense Bed: Distributors, Gas Jets, and Pumping Power

At  $u_0/u_{\rm mf} > 10$  for porous plates and  $u_0/u_{\rm mf} > 3$  for perforated plates, Eq. (9) reduces to

$$\frac{\Delta p_d}{\Delta p_b} \ge \frac{L_f}{L_{\rm mf}} - 1 \tag{10}$$

and if we take  $L_f/L_{\rm mf}=1.2-1.4$ , typical of bubbling beds, Eq. (10) then reduces to  $\Delta p_d \approx (0.2-0.4) \, \Delta p_b$ , which is identical to the rule of thumb given in Eq. (3).

Using large beds (up to 2.4 m square) with tuyere-type distributors, Whitehead et al. [22] carried out extensive experiments with a variety of Celdart B sands. On slowly increasing the gas flow to the bed, they observed that a number of tuyeres became active as soon as  $u_0$  exceeded  $u_{\rm mf}$ , as shown in Fig. 9(a). The number increased progressively until all tuyeres were in use when  $u_0$  reached  $u_1$ . When they reduced the gas flow, all the tuyeres kept operating until a critical velocity  $u_2$  was reached. Then the number of active tuyeres decreased until  $u_{\rm mf}$  was reached. Figure 9(a) shows this hysteresis behavior.

Whitehead et al. also found that  $\Delta p_d$  and  $u_2$  were related to  $L_{\rm in}$ ,  $\rho_z$ , and  $u_{\rm mf}$ . Their final correlation is given in Fig. 9(b), and shows that deeper beds and beds close to  $u_{\rm mf}$  require a larger  $\Delta p_d$  than do shallow beds at high  $u_Q$ .

We summarize these findings with the following design recommendations:

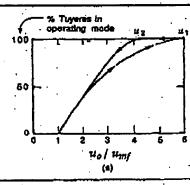
a. For even distribution of fluidizing gas to a bed where  $u_0$  is close to  $u_{\rm inf}$  choose

$$\frac{\Delta p_d}{\Delta p_h} \ge 0.15$$

- b. The required  $\Delta p_d/\Delta p_b$  decreases as  $u_o/u_{\rm mf}$  increases.
- c.  $\Delta p_d/\Delta p_b$  is roughly independent of bed height, or  $\Delta p_b$ .
- d. For the same bed, same uo, and same umi, but different distributor

$$\left(\frac{\Delta p_d}{\Delta p_b}\right)_{\text{porous plate}} > \left(\frac{\Delta p_d}{\Delta p_b}\right)_{\text{orifice plate}}$$

This difference is greater when  $u_0$  is close to  $u_{\rm mf}$  and decreases to zero  $u_0 > u_{\rm mf}$ .



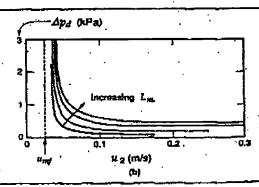


Figure 9. Characteristics of a multibuyere distributor, adapted from Whitehead et al. [22].

Design of Gas Distributor. From:BASELL POLYOLEFINE GMBH, IP +49 69 30527441 07/02/2006 13:33 #309 P.011/012

Design of Gas Distributors

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e. The original rule of thumb

$$\Delta p_d = (0.2-0.4) \, \Delta p_b$$

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is verified by various analyses and experiments and represents a reasonable

upper bound to the required distributor pressure drop for smooth operations.

This value can be made lower in specific cases.

Especially in large-scale commercial-type operations, a severe problem occurs when a distributor is designed for a particular range of operating velocities but is operated in a different range. The problem occurs because  $\Delta p_d$ varies strongly with uo, as given by Eq. (4), whereas  $\Delta p_b$  remains practically independent of uo. So suppose that a distributor is designed according to Eq. (5) for  $u_0 = 2u_{mf}$ . Then at  $u_0 = 20u_{mf}$ , with Eq. (4), we have

$$\frac{\Delta p_d}{\Delta p_b} = \begin{cases} (0.15) \frac{20}{2} = 1.5 & \text{for porous plates} \\ (0.15) \left(\frac{20}{2}\right)^2 = 15 & \text{for perforated plates and tayeres} \end{cases}$$

Such excessive pressure drops could well exceed the capacity of the blowers. Conversely, if a distributor is designed to operate according to Eq. (10) for  $u_0 = 20u_{mf}$ , then, at  $u_0 = 2u_{mf}$ ,  $\Delta p_d$  becomes negligible and the distributor cannot be expected to sustain even fluidization. This discussion suggests that the porous plate distributor can operate satisfactorily over a wider range of gas velocities than can other types of distributors.

Finally, with a properly designed distributor, one without excessive jet penetration, the interaction between the distributor and the bed is limited to a narrow bottom zone of bed. Above this zone, gas-solid contacting is governed primarily by the hydrodynamic properties of the bed itself and cannot be altered

much by changing  $\Delta p_d$ ; see Chap. 5.

Design of Gas Distributors Perforated plates and most trayere distributors can be designed directly from orifice theory, and since the orifice pressure drop is only a small fraction of the total pressure drop, we can use the following procedure.

1. Determine the necessary pressure drop across the distributor,  $\Delta p_d$ , on

the basis of the previous discussion, or simply by using Eq. (3).

2. Calculate the vessel Reynolds number,  $Re_t = d_t u_{\alpha} \rho_{\alpha} / \mu$ , for the total flow approaching the distributor and select the corresponding value for the orifice coefficient, Cd.or.

3. Determine the gas velocity through the orifice, measured at the approach density and temperature:

$$u_{or} = C_{d,or} \left(\frac{2\Delta p_d}{\rho_g}\right)^{1/2} \tag{12}$$

The ratio un un gives the fraction of open area in the distributor plate. See that this is less than 10%.

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OS CHAPTER 4 - The Dense Bed: Distributors, Cas Jets, and Pumping Power

4. Decide on Nor, the number of orifices per unit area of distributor, and find the corresponding orifice diameter from the equation

$$u_o = \frac{\pi}{4} d_{or}^3 u_{or} N_{or} \tag{13}$$

For a tuyers with an inlet orifice, as in Fig. 3(a), Nor should be the number of tuyers per unit area. On the other hand, for tuyers as in Fig. 2(b), but without an inlet orifice (see Fig. 3(b)), Nor is given by

$$N_{or} = \left(\frac{\text{tuyeres}}{\text{area}}\right) \left(\frac{\text{number of holes}}{\text{tuyere}}\right)$$
 (14)

Agitating Distributors. For beds of fine solids such as FCC catalyst, a well-designed distributor should also act as a stirrer, promoting the mixing of solids and keeping the bed well fluidized. The factor that measures this stirring effect is  $\alpha$ , defined as

$$\alpha = \frac{\rho_{\rm g} u_{\rm of}^{\rm g} / 2g_{\rm o}}{\Delta p_{\rm h}} = \frac{\rm kinetic energy of the ordice jets}{\rm resistance of the bed} \tag{15}$$

If a > 1, the jets will punch right through the bed, causing severe gas bypassing, attrition of particles, and erosion of bed internals. If a < 1, the jets will not contribute much to bed stirring, and bubbles rising in the bed will have to do this

What are typical values of a? For a 1-m-high bed of cracking catalyst at ambient conditions, Eq. (3.16) gives, in SI units.

$$\Delta p_b = \frac{(1 - s_{mf})(\rho_s - \rho_g)gL_{mf}}{g_a} = \frac{(1 - 0.4)(1000 - 1)(9.8)(1)}{(1)}$$
= 6000 Fa. (2.60 cm HaQ)

For perforated plate distributors, common practice has  $u_{\rm or}$  ranging between and 60 m/s. Replacing these values in Eq. (15) gives

$$\alpha = \frac{(1.2 \text{ kg/m}^3)(30-60 \text{ m/s})^2}{2(1 \text{ kg-m/s}^2 \text{N})(6000 \text{ Ps})} = 0.09-0.36$$

These values of  $\alpha$  show that gas jets at orifice plates designed for  $u_{or} = 30$  m contribute very little to the stirring of bed solids. At  $u_{or} = 60$  m/s, or reasonable stirring by the distributor jets can be expected; nevertheless, friable particles one may prefer to select the distributor plate with smaller a

Avoidance of High let Velocities at Distributors. When a high velocity cannot be used to provide the needed distributor pressure drop because of particle attrition, then one must use inlet orifices for the tryeres, as shown Fig. 3(a). For orifice plates, one can meet these requirements by proposelecting  $N_{\rm or}$  and  $d_{\rm or}$ .

EXAMPLE 1

Design a perforated plate distributor for use in a commercial fluidized bed re-

Date

$$d_t = 4 \text{ m}$$
  $l_{mf} = 2 \text{ m}$   $s_{mf} \approx 0.48$   
 $\rho_0 = 1500 \text{ kg/m}^3$   $\rho_0 = 3.6 \text{ kg/m}^3$   $\mu = 2 \times 10^{-8} \text{ kg/m}$ 

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